



## Intake of Inorganic Arsenic in the North American Diet

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### ABSTRACT

Dietary intake of inorganic arsenic, previously assumed to be an insignificant source of arsenic exposure in humans, was estimated for Canadian and United States populations. Input data included arsenic contents of various food groups, a limited historical database from the Ontario Ministry of the Environment measuring the percent inorganic arsenic in food groups, and food consumption data. Estimated daily dietary intake of inorganic arsenic ranges from 8.3 to 14 µg/day in the United States and from 4.8 to 12.7 µg/day in Canada for various age groups. These data suggest that between 21% to 40% of total dietary arsenic occurs in inorganic forms. Uncertainties regarding total arsenic in dairy products in the data set applied here may account for observed differences between United States and Canadian estimates. While estimates provided here are preliminary because of limitations in data on the proportion of inorganic arsenic in foods, this analysis suggests that dietary intake of inorganic arsenic is higher than is currently assumed. Additional research is needed to more fully characterize inorganic arsenic concentrations in foods. Future study is also needed on the variability of total and inorganic arsenic in foods and the bioavailability of dietary inorganic arsenic.

**Key Words:** United States, Canada, food, speciation, metals exposure assessment, arsenic

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## INTRODUCTION

As the 20th most abundant element in the Earth's crust, arsenic has been detected in virtually all foods evaluated (NAS, 1977; Irgolic, 1992). Speciation analyses of arsenic have focused primarily on marine animals; less data are available for marine plants and less yet are available for terrestrial biota and foods as consumed (Irgolic, 1992; Phillips, 1994). (A separate manuscript [Schoof *et al.*, 1998] has been submitted presenting results of speciation analyses of arsenic in yams and rice collected in Taiwan. The results of these analyses are new data that can be used to evaluate arsenic concentrations in yams and rice.) Arsenic in marine biota has been shown to occur predominantly in nontoxic organic forms (*i.e.*, arsenobetaine and arsenocholine). Because of the widespread belief that most dietary arsenic also occurs in nontoxic organic forms, dietary intake of inorganic arsenic is typically considered to be insignificant (Gunderson, 1995). This may not be the case.

A preliminary study of speciated arsenic in food was conducted by the Ontario Ministry of the Environment (OME). Results have been circulated in internal memoranda (OME, 1987), but they have never been published. A review of the OME (1987) data in light of reports evaluating the data shows that these data have been widely misinterpreted because of an inaccurate table in a widely cited U.S. Environmental Protection Agency (EPA) report (USEPA, 1988).<sup>1</sup> For example, although OME did not analyze any potatoes or vegetables, the data have been cited as indicating that arsenic in yams (USEPA, 1988) and vegetables (Mushak and Crocetti, 1995) occurs primarily in organic forms. This paper presents the findings of the 1987 OME study and applies them to provide preliminary estimates of inorganic arsenic intake in typical United States and Canadian diets.

## SPECIATION ANALYSES CONDUCTED BY OME

To identify the relative proportions of inorganic and organic arsenic in foods, OME analyzed 15 samples of food for total arsenic and for inorganic and organic arsenic forms. These analyses were carried out by the Ontario

<sup>1</sup> Table E-1 of USEPA (1988) cites the OME study presented here as the source of estimates of the percentage of inorganic arsenic in food groups that EPA applied in deriving toxicity values for arsenic. None of the EPA inorganic arsenic percentages match those detected by OME. EPA estimates for saltwater fish (0%), rice (35%), and cereals (65%) are close to those detected by OME (*i.e.*, EPA estimates are within 5–10 percentage points of those predicted by OME data). However, EPA estimates for milk (75%) and poultry (65%) appear to be the inverse of those detected by OME for milk (26%) and poultry (41%), EPA estimates for fruit (10%) does not agree with the percent suggested by OME data (73%), and there were no OME analyses for potatoes or vegetables and thus these EPA estimates (of 90% and 95%, respectively) are unexplained.

Research Foundation (ORF) for the OME. Method development was reported in one technical memorandum (OME, 1986) and results in a second (OME, 1987).<sup>2</sup>

### Methods

Total and speciated arsenic were measured in 15 homogenized food samples (OME, 1986). Although only one sample was analyzed for each food, all but four of the food samples were analyzed in duplicate or triplicate (i.e., all except sole, tuna, apple juice, and cigarettes).

For total arsenic measurements, aliquots of 1 to 7 g (depending on moisture and fat content) were measured into 125-ml flasks, 20 ml concentrated nitric acid was added, and samples were warmed until the initial reaction subsided. Sulfuric acid (2 ml) was then added, and the solutions were evaporated to light fumes of sulfuric acid. When necessary to prevent charring or loss of sample, nitric acid was added. Perchloric acid (2 ml) and nitric acid (5 ml) were then added, the mixture was evaporated to destroy residual organic material and expel remaining perchloric acid, and samples were allowed to cool. Deionized water (10 ml) and hydrochloric acid (5 ml) were then added. The solutions were warmed to dissolve precipitated salts, cooled, and diluted to volume (25 ml). Finally, the solutions were reduced from the pentavalent to the trivalent state using potassium iodide and analyzed for total arsenic by hydride atomic absorption.

Analyses for speciated arsenic began by digesting subsamples of the foods analyzed for total arsenic using hydrochloric acid (25 ml of a 50% solution) and hydrobromic acid (1 ml) and then refluxing samples in a Bethge distillation apparatus for 5 to 15 minutes until 20 ml of distillate could be collected. Then an additional 20 ml of hydrochloric acid was added, and 20 ml more of the distillate was collected. Condensers and receivers were rinsed, and the rinsate was added to the combined distillate.

Inorganic arsenic was reduced to the trivalent state during distillation and codistilled with the acid mixture. Distillates containing the inorganic arsenic were combined with nitric acid (5 ml) and sulfuric acid (2 ml), and the solutions were evaporated to fumes of sulfuric acid. After cooling, water (10 ml) and hydrochloric acid were added and the solutions were diluted to 25 ml for hydride generation atomic absorption analysis.

Organic arsenic was determined by taking the residues in the distillation flask: adding concentrated nitric (approximately 20 ml), sulfuric (2 ml), and perchloric (5 ml) acids; evaporating to fumes of perchloric acid; diluting; and detecting with hydride generation atomic absorption. OME (1986) mentions possible breakdown of organic arsenic during the distillation step and notes that evaporating the distillation flask to dryness could cause further decomposition of organic compounds.

<sup>2</sup> The principal investigator, Roland Weiler, has retired and could not be contacted, consequently, some details of the procedures are unknown.

## Results

Total arsenic concentrations in the foods analyzed ranged from 0.011 mg/kg in pastry flour to 4 mg/kg in sole (Table 1; all sample results are reported as wet weight, except as indicated). Inorganic arsenic concentrations ranged from 0.0042 mg/kg in vanilla ice cream to 0.1 mg/kg in rice and shrimp, and organic arsenic concentrations ranged from undetected in a variety of foods to 0.52 mg/kg in canned shrimp. The percent inorganic arsenic in these foods, calculated here by dividing the average inorganic arsenic for a specific food by the total arsenic for that food,<sup>3</sup> ranged from 1% for marine fishes to 100% for meat (based on samples of pork and pastrami). When data were available for several foods from a food group (*i.e.*, in the case of meat, saltwater fish, and cereals), the average for that food group was also calculated (Table 1).

## INTAKE OF INORGANIC ARSENIC

Methods used in estimating dietary intake of inorganic arsenic in United States and Canadian populations are described in the following sections.

### United States Diet

Total arsenic intake from a typical diet in the United States was calculated from data compiled by the U.S. Food and Drug Administration (FDA) on food consumption patterns and total arsenic concentrations in foods. Food consumption patterns for United States populations were based on FDA market basket surveys for 1982 through 1990. These surveys provide consumption rates for 11 general food groups that represent the diets of United States populations in three age categories: infants (0 to 6 months), toddlers (6 months to 2 years), and adults (18 years and older) (Borum, 1992; Gunderson, 1995). FDA also reports total arsenic concentrations detected in foods that correspond with the categories evaluated in the consumption surveys (Gunderson, 1995). Foods were prepared for cooking, cooked, digested with nitric, perchloric, and sulfuric acids, and analyzed with hydride generation atomic absorption. In a background document prepared by EPA (Borum, 1992), food consumption data were combined with FDA measurements of total arsenic concentrations in foods to estimate total arsenic intakes of 21.5 µg/day for infants, 27.6 µg/day for toddlers, and 52.6 µg/day for adults.

To derive the inorganic arsenic intake estimates in Table 2, the FDA's total arsenic estimate for each food group presented in Borum (1992) was multiplied by the OME estimates of the percent inorganic arsenic for the corresponding food groups (Table 1). Certain FDA categories did not have an exact counterpart in the OME (1987) study. Specifically, no OME data were avail-

<sup>3</sup> For some samples, less than 100% of the total arsenic was recovered as inorganic arsenic. We calculated inorganic arsenic as a percent of total arsenic, based on the assumption that unrecovered arsenic was either in complex organic forms, or if present as inorganic arsenic, it would not be bioavailable.

TABLE 1. SPECIATED ARSENIC DATA FROM ONTARIO  
MINISTRY OF THE ENVIRONMENT<sup>a</sup>

Food Category	(N)	Arsenic Concentration			% Inorganic Arsenic <sup>b</sup>
		Total	Inorganic	Organic	
Milk and Dairy Products (average)					26
Vanilla ice cream (average of replicates)	1	0.016	0.0042	<0.002	26
First replicate			0.0035	<0.001	
Second replicate			0.0049	<0.002	
Meat (average)					100
Pork (cured, average of replicates)	1	0.013	0.018	<0.007	144
First replicate		0.013	-	-	
Second replicate		0.012	-	-	
Pastrami (average of replicates)	1	0.024	0.024	<0.005	95
First replicate		0.023	-	-	
Second replicate		0.024	-	-	
Third replicate		0.026	-	-	
Poultry (average)					41
Chicken (average of replicates)	1	0.022	0.0090	0.012	41
First replicate		0.021	-	-	
Second replicate		0.023	-	-	
Fish (saltwater) (average)		2.55	0.024	2.8	1
Sole	1	4	0.022	4.4	1
Tuna	1	1.1	0.025	1.2	2
Fish (freshwater) (average)					15
Pickering (average of replicates)	1	0.14	0.022	0.086	15
First replicate		-	0.019	-	
Second replicate		-	0.024	-	

TABLE 1. (cont.)

Food Category	(N)	Arsenic Concentration			% Inorganic Arsenic <sup>a</sup>
		Total	Inorganic	Organic	
Shellfish					16
Shrimp (average of replicates)	1	0.6E	0.10	0.5E	1E
First replicate		-	0.12	-	
Second replicate		-	0.086	-	
Rice (average of replicates)	1	0.24	0.1	0.1E	43
First replicate		0.24	0.1	-	
Second replicate		0.23	0.1	-	
Cereals (average for all)					49
"Special K" (average of replicates)	1	0.27	0.070	0.1E	2E
First replicate		0.3	-	-	
Second replicate		0.23	-	-	
Bread (whole wheat, average of replicates)	1	0.024	0.012	<0.006	50
First replicate		-	0.011	-	
Second replicate		-	0.012	-	
Pastry flour (average of replicates)	1	0.011	0.007E	<0.00E	6E
First replicate		0.011	-	-	
Second replicate		0.011	-	-	
Fruit					73
Apple juice	1	0.012	0.008E	<0.002	73
Vegetables		-	-	-	NA
Potatoes		-	-	-	NA

TABLE 1. (cont.)

Food Category	(N)	Arsenic Concentration			% Inorganic Arsenic <sup>b</sup>
		Total	Inorganic	Organic	
Tee (average of replicates)	1	0.035	0.0091	0.025	26
First replicate		0.035	0.0091	0.025	
Second replicate		-	<0.02	<0.02	
Cigarettes	1	0.15	0.11	0.02	61

Note: Values expressed as mg/kg wet weight.

- not analyzed

OME - Ontario Ministry of the Environment

<sup>a</sup> Arsenic concentration data are reproduced from OME (1987). Percent recovery and percent inorganic arsenic were calculated by the present authors.

<sup>b</sup> Estimated from OME data by dividing inorganic arsenic concentration by total arsenic concentrations. Boxed values represent the entire food group, other values represent individual foods.

TABLE 2. DIETARY INTAKE OF ARSENIC IN  
UNITED STATES AND CANADIAN POPULATIONS

Food Category	Children					
	U.S. Diet <sup>a</sup>				Canadian Diet <sup>b</sup>	
	Infant		Toddler		Ages 1-4	
	Total	Inorganic	Total	Inorganic	Total	Inorganic
Dairy	13.4	3.5	8.5	2.2	1.1	0.3
Meat	0.6	0.6	0.5	0.5	0.9	0.5
Poultry	0.4	0.1	1.1	0.4	0.6	0.3
Fish (saltwater)	0.0	0.0	6.0	0.1	5.7	0.1
Fish (freshwater)	0.0	0.0	0.4	0.1	0.6	0.1
Shellfish	0.0	0.0	0.5	0.1	0.6	0.1
Legumes <sup>c</sup>	1.2	0.6	0.4	0.2	0.2	0.1
Rice <sup>c</sup>	0.3	0.1	1.1	0.5	0.7	0.3
Cereals	1.7	0.8	2.5	1.2	1.8	0.9
Fruit	2.1	1.6	1.9	1.4	1.1	0.8
Vegetables <sup>c</sup>	1.2	0.6	1.2	0.6	0.3	0.1
Potatoes <sup>c</sup>	0.2	0.1	0.5	0.4	0.6	0.4
Tea	NA	NA	NA	NA	0.0	0.0
Other foods <sup>d</sup>	0.5	0.2	2.6	1.3	0.7	0.4
<b>Totals</b>	<b>21.5</b>	<b>8.3</b>	<b>27.6</b>	<b>9.4</b>	<b>15.1</b>	<b>4.8</b>
<b>Totals without dairy<sup>e</sup></b>	<b>8.1</b>	<b>4.7</b>	<b>19.1</b>	<b>7.1</b>	<b>14.0</b>	<b>4.5</b>



TABLE 2. (cont.)

Food Category	Adults					
	U.S. Diet <sup>a</sup>		Canadian Diet			
			Women 20-35		Men 20-35	
	Total <sup>b</sup>	Inorganic	Total	Inorganic	Total	Inorganic
Dairy	4.7	1.2	0.6	0.2	0.9	0.2
Meat	2.6	2.6	2.1	2.1	3.5	3.5
Poultry	2.1	0.5	1.2	0.5	1.6	0.7
Fish (saltwater)	23.9	0.3	17.5	0.2	33.3	0.6
Fish (freshwater)	1.5	0.2	0.3	0.1	1.0	0.1
Shellfish	1.5	0.3	3.5	0.6	6.2	1.0
Legumes <sup>c</sup>	0.6	0.3	0.2	0.1	0.3	0.2
Rice <sup>c</sup>	1.3	0.5	1.6	0.7	1.5	0.8
Cereals	3.1	1.5	2.4	1.2	3.5	1.7
Fruit	1.7	1.3	0.8	0.6	1.0	0.6
Vegetables <sup>c</sup>	3.3	1.5	0.7	0.3	0.8	0.4
Potatoes <sup>c</sup>	1.2	0.5	0.7	0.5	1.4	1.0
Tea	NA	NA	0.6	0.2	0.6	0.2
Other foods <sup>d</sup>	4.7	2.4	1.8	0.9	3.6	1.8
Totals	52.6	14.0	34.4	8.1	59.6	12.7
Totals without dairy <sup>e</sup>	47.5	12.7	33.8	7.9	58.6	12.5

Footnotes on next page.

TABLE 2. (cont.)

Note: Values expressed as  $\mu\text{g/day}$ .

FDA	- U.S. Food and Drug Administration
NA	- no data available
OME	- Ontario Ministry of the Environment

<sup>a</sup> Estimates based on percentages of inorganic arsenic from OME (unless otherwise noted) combined with FDA market basket consumption values for 1982 to 1990 and total arsenic concentrations as reported in Borum (1992). Infants are up to 6 months, toddlers are 6 months to 2 years, and adults are 18 years and older.

<sup>b</sup> Estimates based on percentages of inorganic arsenic from OME (unless otherwise noted) combined with total arsenic concentrations from the Canadian Health Protection Branch and intake from Nutrition Canada as reported in Dabeka et al. (1993). Totals for each food group in Dabeka et al. (1993) were used in these estimates and the overall sum for all foods does not match the overall sum in Dabeka et al. (1993).

<sup>c</sup> No legumes or vegetables were measured by OME: 47% inorganic arsenic assumed (average of rice and cereals in OME).

<sup>d</sup> FDA category for "mixture mainly grain" used in estimates for U.S. populations.

<sup>e</sup> No potatoes were analyzed by OME: 75% inorganic arsenic assumed based on the average inorganic arsenic detected in yams in Schoof et al. (1997).

<sup>f</sup> Other foods were assumed to contain 50% inorganic arsenic, based on the average of all foods analyzed by OME of 46%.

<sup>g</sup> Concentrations of total arsenic in dairy in U.S. populations are uncertain because of a high number of undetected values in dataset. See text.

TABLE 3. COMPARISON OF TOTAL ARSENIC DETECTED IN

OME AND DABEKA *et al.* (1993)

Food Category	OME <sup>a</sup>	Dabeka <i>et al.</i> (1993) <sup>b</sup>		OME/Dabeka <i>et al.</i> (1993)
	Average	Average	Range	Average
Milk and Dairy Products (average)	-	0.003E	0.004-0.02E	-
Vanilla ice cream	0.01E	0.00E	0.0007-0.010	3.2
Meat (average)	-	0.02E	<0.001-0.53E	-
Pork (cured)	0.01E	0.01E	0.0081-0.02E	0.7
Pastrami	0.024	0.014	0.0062-0.037	1.7
Poultry (average)	-	0.02E	<0.001-0.1	-
Chicken	0.022	0.047	0.018-0.1	0.5
Fish (saltwater) (average)	2.550	3.0E	1.85-4.8E	0.8
Fish (freshwater) (average)	0.14E	0.4E	0.077-1.3E	0.3
Shellfish	0.65E	2.04	1.01-4.2	0.3
Rice (average)	0.23E	0.097	0.075-0.38E	2.4
Cereals (average)	0.10E	0.011	<0.0001-0.14E	9.5
Fruit (apple juice)	0.012	0.0060	0.0045-0.0094	2.0
Vegetables	-	0.0053	<0.0001-0.03E	-
Potatoes	-	0.09E	<0.0001-0.044	-
Ter	0.03E	0.0021	0.0004-0.0051	16.7

Note: Values express as mg/kg wet weight.

All values are averages of replicates or averages of foods in the group except apple juice, which is a single value.

- - data not available

OME - Ontario Ministry of the Environment

<sup>a</sup> See Table 2 for specific products from these food categories analyzed by OME.

<sup>b</sup> Foods were selected from Dabeka *et al.* (1993) that most closely approximated foods analyzed by OME.

able for legumes or vegetables; an estimate of 47% inorganic arsenic was applied to these foods based on the average percentage of inorganic arsenic detected in rice and cereals. Because no OME data were available for potatoes, the estimate of average inorganic arsenic in yams of 76% from Schoof *et al.* (1998) was applied here. The FDA category of "other foods," which included data for oils, beverages (other than those prepared from dairy or fruit products), coffee, and additional foods, was assumed to contain 50% inorganic arsenic based on the average of the OME percentages for all food categories analyzed. These calculations yielded total United States dietary intake estimates for inorganic arsenic of 8.3  $\mu\text{g}/\text{day}$  for infants, 9.4  $\mu\text{g}/\text{day}$  for toddlers, and 14.0  $\mu\text{g}/\text{day}$  for adults (Table 2). Intake estimates that exclude arsenic intake from dairy products are also presented in Table 2 because of uncertainties in the estimates for that food category (see *Discussion* below).

#### Canadian Diet

Total arsenic intake in the typical Canadian diet was reported by Dabeka *et al.* (1993), who summarized age- and sex-specific consumption rate data for 112 food categories representative of the Canadian diet and corresponding total arsenic concentrations. Consumption data were collected by the Nutrition Canada Survey of the Canadian Department of Health and Welfare. Total arsenic concentrations in food samples from the 112 food categories were collected from six Canadian cities and compiled by the Canadian Total Diet Program. Food samples had been prepared for consumption, homogenized, and then digested in nitric acid prior to measurement of total arsenic by graphite furnace atomic absorption.

The intake of total arsenic averaged over the six Canadian cities ranged from 15.1  $\mu\text{g}/\text{day}$  for children ages 1–4 to 59.6  $\mu\text{g}/\text{day}$  for adult men ages 20–39. The overall average for the entire population was 38.5  $\mu\text{g}/\text{day}$  (Dabeka *et al.*, 1993).

To derive the inorganic arsenic estimates in Table 2, the total arsenic concentrations for the food categories reported in Dabeka *et al.* (1993) were multiplied by our estimates of percent inorganic arsenic in the corresponding groups (Table 1). These calculations yielded estimates of total Canadian dietary intake of inorganic arsenic of 4.8  $\mu\text{g}/\text{day}$  for children ages 1–4, 8.1  $\mu\text{g}/\text{day}$  for women ages 20–39, 12.7  $\mu\text{g}/\text{day}$  for adult men ages 20–39, and 8.3  $\mu\text{g}/\text{day}$  for all ages combined.

#### DISCUSSION

The analyses presented in this paper suggest that inorganic arsenic comprises approximately 20% to 40% of total dietary arsenic intake. Additional research is needed to confirm these estimates. Cereals, rice, and fish, identified as relatively important sources of total arsenic in the diet (Dabeka *et al.*, 1993; Gunderson, 1995), were well characterized in the OME data set and appear to be important sources of inorganic arsenic as well. The estimates for rice and fish are also supported by other studies. Schoof *et al.* (1998) found an

average of 68% inorganic arsenic in speciation analyses of seven rice samples. Norin *et al.* (1985) reported 5% to 22% inorganic arsenic in freshwater fish, and a review by Shiomi (1994) reported inorganic arsenic in saltwater fish ranging from 0% to 3%.

Additional inorganic arsenic data on other relatively large dietary arsenic sources such as dairy products (*i.e.*, milk with a range of fat content), beef (*i.e.*, hamburger or steak), poultry (*i.e.*, eggs), and potatoes would reduce uncertainties associated with the use of the OME data set. Although fruits and vegetables do not appear to be primary contributors to total dietary arsenic intake, information on the proportion that is inorganic would be useful in conducting human health risk assessments where consumption of homegrown produce is often evaluated. While arsenic intake from home-grown produce is often dismissed as being irrelevant because only nontoxic organic forms are present, these limited data suggest further evaluation is warranted.

Data in Table 2 suggest United States dietary intake of inorganic arsenic is higher than Canadian intake. The most comparable age categories between the two data sets are United States toddlers (6 months to 2 years old), with a daily inorganic arsenic intake of 9.4  $\mu\text{g}$ , and 1–4 year old Canadians, with a daily intake of 4.8  $\mu\text{g}$ . Although the inorganic arsenic intake estimate for United States adults, 14.0  $\mu\text{g}/\text{day}$ , was similar to that for Canadian men ages 20–39, 12.7  $\mu\text{g}/\text{day}$ , the estimate for United States adults was higher than estimates for Canadian women ages 20–39 (8.1  $\mu\text{g}/\text{day}$ ). The single largest difference in intake of inorganic arsenic between United States and Canadian populations appears to be from milk and dairy products. United States intake values range from 1.2–3.5  $\mu\text{g}/\text{day}$ , while intake in Canadian populations ranges from 0.2–0.3  $\mu\text{g}/\text{day}$  (Table 2). This categorical difference appears to be large enough to account for most of the overall difference in intake (Table 2).

The summary of FDA data used in the current estimates (Borum, 1992) did not allow an exact comparison of consumption rates for dairy products in United States and Canadian populations. A summary of United States consumption rates estimated from 1980–1982 FDA data (Gartrell *et al.*, 1986) suggests, however, that differences in intake from dairy products may be related to higher consumption rates of these products in United States populations. Gartrell *et al.* (1986) reported higher average daily consumption rates of dairy products for United States adults (761 g) than were reported by Dabeka *et al.* (1993) for all Canadians (442 g), Canadian women ages 20–39 (291 g), or Canadian men ages 20–39 (425 g). Verification of summary data on consumption rates for dairy products in United States populations would be useful because this food category is an important contributor of dietary arsenic; however, consumption rate differences alone do not appear to be sufficient to explain the differences in the two data sets.

Observed differences in intake of arsenic from dairy products may be due to uncertainties in the total arsenic concentrations derived from the FDA summary used here (Borum, 1992) where total arsenic concentrations were derived from the average of the detected sample, excluding any nondetected

samples from the estimate. Because there were very few total arsenic detections in dairy products, total arsenic averages based solely on detected samples may have resulted in an overestimate of total arsenic concentrations. Data sets for other food groups had a much smaller proportion of nondetected samples and are less likely to be overestimates. Estimates derived by Dabeka *et al.* (1993) used detection limits in calculating averages including nondetected samples and thus, these estimates are also less likely to overestimate concentrations of total arsenic in dairy products. Calculations of inorganic arsenic intake excluding intake from dairy products show much closer agreement between United States and Canadian populations (Table 2).

Application of inorganic arsenic data from the 15 individual food types analyzed by OME to derive estimates of inorganic arsenic intake for all foods provides preliminary estimates that need to be confirmed by additional studies. Variability of total arsenic among foods within a food group (*e.g.*, specific dairy products within the milk and dairy food group) analyzed by Dabeka *et al.* (1993) often spans an order of magnitude, while data from OME are only available for one food each from the dairy, fruit, and poultry categories. In addition, some food groups (*e.g.*, legumes, potatoes, vegetables) were not represented among the food samples analyzed by OME; the use of extrapolated values from other food groups to represent these food groups may under- or overestimate inorganic arsenic exposures.

Very few studies of arsenic forms in food have been performed, and concerns have been raised that the strong acid digestions used in analyses of organic and inorganic arsenic could break down organic arsenic compounds (Mushak and Crocetti, 1995). Nevertheless, virtually all the speciated arsenic recovered in fish was present in organic forms (*i.e.*, the average percent organic arsenic concentration was 99.6% in two marine fish samples and one shrimp sample) (Table 1). This issue is discussed in more detail in Slayton *et al.* (1996) and in Schoof *et al.* (1998).

Although OME analyzed a limited number of individual foods, the availability of replicate samples and the relatively good agreement of replicates strengthens the OME findings. Eleven of the 15 foods analyzed by OME were analyzed in duplicate or triplicate for either total or inorganic arsenic, with percent differences in replicates ranging from 0% to 29% (Table 1). The good agreement between total arsenic concentrations reported by OME and by Dabeka *et al.* (1993) also suggests that the OME data set accurately represents arsenic concentrations. Average total arsenic concentrations measured by OME are generally within a factor of three of average concentrations measured for those foods by Dabeka *et al.* (1993). Total arsenic detected in all but three of the foods analyzed by OME were within the range reported by Dabeka *et al.* (1993) (Table 3).<sup>4</sup>

<sup>4</sup> At the time of preparation of this manuscript, the authors did not have data needed to conduct a similar comparison of total arsenic in specific foods analyzed by FDA.

Risk assessment of ingested arsenic is based on toxicity values derived from a population exposed to arsenic in drinking water. Absorption of arsenic from soil is less complete than the absorption of arsenic from water (Freeman *et al.*, 1995; Groen *et al.*, 1994; Ruby *et al.*, 1996). Arsenic in food may also have limited bioavailability. To accurately estimate exposure to inorganic arsenic in food, it is necessary to determine the absorption of arsenic from dietary sources. Research on the bioavailability of arsenic in food samples would provide data to more accurately assess the importance of dietary arsenic intake.

## CONCLUSIONS

The OME data set published here was used by EPA as a basis for estimates of dietary intake of inorganic arsenic used in developing toxicity values for ingested arsenic. While estimates provided here are preliminary, because of limitations in the OME data set and uncertainties in total arsenic in dairy products in the United States, this analysis suggests that dietary intake of inorganic arsenic is higher than previously assumed. Additional research is needed to more fully characterize inorganic arsenic concentrations in food types. Future study is also needed on the variability of total and inorganic arsenic in foods and the bioavailability of dietary inorganic arsenic.

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